

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re the application of:

Kenji OKADA, et al.

Group Art Unit: 1796

Serial Number: 10/594,228

Examiner: Sanza L. McClendon

Filed: September 25, 2006

For: COMPOSITION CURABLE BY RADICAL PHOTO CURING AND  
CATIONIC PHOTO CURING IN COMBINATION

DECLARATION UNDER 37 CFR 1.132

Commissioner for Patents

P.O. Box 1450

Alexandria, Virginia 22313-1450

Sir:

Yoshikatsu Ichiryu, citizen of Japan, duly deposes and says:

1. That he has graduated from Faculty of Engineering of Kansai University, Japan, in the year of 2000, and he received the degree of Master of Applied Chemistry from Kansai University, Japan in the year of 2002;

2. That he has been employed in his capacity since 2002 by KANEKA CORPORATION;

3. That he has been engaged in research and development on high performance polymers;

4. That he is familiar with the instant application for United States Letters Patent and the Office Action thereto mailed October 5, 2009; and

5. That he conducted the experiments described below, in which in order to prove surprising and unexpected results achieved by the amended claim 1 of the present invention, the compositions and the

cured articles of the amended claim 1 obtained by employing an epoxy compound together with applying ultraviolet curing are superior to those obtained by the method corresponding to Example 9 in Nakagawa et al. (US 6,964,999) in low viscosity of the composition and mechanical properties of the cured article.

### **Experiments**

The object of the experiments is to prove that low viscosity of the composition and improved mechanical properties of the cured article of the present invention are achieved by employing an epoxy compound together with applying ultraviolet curing, by evaluating mechanical properties such as modulus (M30) at 30% elongation, strength (Tb) at break and elongation (Eb) at break of the cured article obtained by the method corresponding to Example 9 in Nakagawa et al. (US 6,964,999), and by comparing the composition of Nakagawa et al. with those employed in Examples and Comparative Example of the instant application.

Following Examples 1 and 2 are identical to Examples 1 and 4 of the instant application respectively, and following Comparative Example 1 is identical to Comparative Example 1 of the instant application. Following Comparative Example 2 corresponding to Example 9 in Nakagawa et al. (US 6,964,999) is newly conducted.

### **Production Example**

N-butyl acrylate/ethyl acrylate/2-methoxyethyl acrylate were polymerized at a ratio of 25/46/29 by mole using cuprous bromide as a catalyst, pentamethyldiethylenetriamine as a ligand and diethyl 2,5-dibromoadipate as an initiator to obtain poly(n-butyl acrylate/ethyl acrylate/2-methoxyethyl acrylate) having a number average molecular weight of 16,500, a molecular weight distribution of 1.13 and bromine

groups at its ends.

400 g of the polymer was dissolved in N,N-dimethylacetoamide (400 ml), 10.7 g of potassium acrylate was added and the mixture was heated with stirring at 70°C for 6 hours under nitrogen atmosphere to obtain a mixture composed of poly(n-butyl acrylate/ethyl acrylate/2-methoxyethyl acrylate) having acryloyl groups at both ends. After the N,N-dimethylacetoamide in the mix solution was removed under reduced pressure, toluene was added to the residue and the insoluble portion was removed by filtration. The toluene in the filtrate was removed under reduced pressure to give a purified polymer.

With respect to the above-mentioned polymer having acryloyl groups at both ends after purification, the number average molecular weight was 16,900, the molecular weight distribution was 1.14 and the average number of end acryloyl group was 1.8 (namely, the introduction percentage of the acryloyl group to each end was 90 %).

#### Example 1

0.20 Part by weight of 2,2-diethoxyacetophenone, 30 parts by weight of EPOLIGHT 4000 (a hydrogenated bisphenol A epoxy resin available from KYOEISHA CHEMICAL Co., Ltd.), 1.5 parts by weight of OPTOMER SP172 (a cationic photopolymerization initiator available from Asahi Denka Co., Ltd.) (5 parts by weight based on EPOLIGHT 4000) and 1 part by weight of Irganox 1010 [pentaerythritol tetrakis[3-(3,5-di-t-butyl-4-hydroxyphenyl)propionate] available from Chiba Specialty Chemicals Co., Ltd.] were added to 100 parts by weight of the polymer obtained in Production Example and the mixture was adequately mixed to obtain a curable composition.

Then, the curable composition in a sheet form was passed 5 times with respect to each of front and rear surfaces thereof under a metal halide lamp (80W/cm, an irradiation distance of 15 cm and a belt speed of 1.0 m/min) for light irradiation to obtain a sheet-like cured article with a thickness of about 2 mm.

The viscosity of the curable composition and the mechanical properties of the cured article were measured by the following methods:

[Viscosity measurement]

The viscosity of the curable compositions prepared in Examples and Comparative Examples was measured under the conditions of a measurement temperature of 23°C and a cone of 3° using an E type viscometer (VISCONIC ED type; manufactured by Tokyo Keiki Co., Ltd.) according to JIS K 7117-2.

[Evaluation of mechanical properties]

The mechanical properties (modulus (M30) at 30% elongation, strength (Tb) at break and elongation (Eb) at break) of the cured articles prepared in Examples and Comparative Examples were measured for dumbbell No.2 specimen using an autograph AG-2000A manufactured by Shimadzu Corporation according to JIS K 7113.

Example 2

0.20 Part by weight of 2,2-diethoxyacetophenone, 100 parts by weight of EPOLIGHT 4000, 5 parts by weight of OPTOMER SP172 (5 parts by weight based on EPOLIGHT 4000) and 1 part by weight of Irganox 1010 were added to 100 parts by weight of the polymer obtained

in Production Example and the mixture was adequately mixed to obtain a curable composition.

Then, the curable composition in a sheet form was passed 5 times with respect to each of front and rear surfaces thereof under a metal halide lamp (80W/cm, an irradiation distance of 15 cm and a belt speed of 1.0 m/min) for light irradiation to obtain a sheet-like cured article with a thickness of about 2 mm.

The viscosity of the curable composition and the mechanical properties of the cured article were measured.

#### Comparative Example 1

0.20 Part by weight of 2,2-diethoxyacetophenone and 1.0 part by weight of Irganox 1010 were added to 100 parts by weight of the polymer obtained in Production Example and the mixture was adequately mixed to obtain a curable composition.

Then, the curable composition in a sheet form was passed 3 times under a metal halide lamp (80W/cm, an irradiation distance of 15 cm and a belt speed of 1.0 m/min) for light irradiation to obtain a sheet-like cured article with a thickness of about 2 mm.

The viscosity of the curable composition and the mechanical properties of the cured article were measured.

#### Comparative Example 2

0.20 Part by weight of 2,2-diethoxyacetophenone was added to 100 parts by weight of the polymer obtained in Production Example and the mixture was adequately mixed to obtain a curable composition.

Then, the curable composition in a sheet form was passed 3 times under a metal halide lamp (80W/cm, an irradiation distance of 15

cm and a belt speed of 1.0 m/min) for light irradiation to obtain a sheet-like cured article with a thickness of about 2 mm.

The mechanical properties of the cured article were measured.

## **RESULTS AND DISCUSSIONS**

The viscosity of the compositions and the mechanical properties of the cured articles measured in the experiments described above are shown in Table 1.

TABLE 1

	Ex.1	Ex.2	Com.Ex.1	Com.Ex.2
Composition				
Polymer	100	100	100	100
EPOLIGHT 4000	30	100	-	-
OPTOMER SP172	1.5	5.0	-	-
2,2-diethoxyacetophenone	0.2	0.2	0.2	0.2
Irganox 1010	1.0	1.0	1.0	-
Viscosity (23°C)[Pa×s]	150.0	43.0	510	-
Mechanical properties				
M30 (MPa)	0.52	4.39	0.26	0.33
Tb (MPa)	0.94	5.33	0.48	0.51
Eb (%)	52	37	59	50

As is apparent from the results shown in Table 1, the cured article employed in Comparative Example 2 was equivalent to that employed in Comparative Example 1 in the mechanical properties (M30,

Tb and Eb), and Comparative Example 1 is identical to Comparative Example 2 except addition of antioxidant (Irganox 1010).

Therefore, it is understood that the composition in Comparative Example 2 shows similar viscosity to the composition in Comparative Example 1. In other words, it is thought that the viscosity of the composition in Example 9 of Nakagawa et al. falls into around 510 Pa·s.

By contrast, the mechanical properties of the cured articles in Examples 1 to 2 employing an epoxy compound together with applying ultraviolet curing showed M30 of 0.52 MPa and 4.39 MPa respectively. Those are at least approximately two times higher values as compared with those of the cured articles in Comparative Examples 1 and 2 as well as Tb. The results clearly show that the cured article obtained by employing an epoxy compound together with applying ultraviolet curing is excellent in M30 and Tb, and also show that the cured article obtained by employing an epoxy compound together with applying ultraviolet curing is equivalent to that obtained by not employing an epoxy compound together with ultraviolet curing with respect to Eb.

In addition, the viscosities of the composition in Examples 1 and 2 were 150.0 Pa·s and 43.0 Pa·s respectively. These results are at least 3 times lower values as compared with those in Comparative Examples. The results clearly show that the composition employing an epoxy compound together with applying ultraviolet curing is excellent in low viscosity of the composition.

Accordingly, it is proved that the composition and the cured article obtained by employing an epoxy compound together with applying ultraviolet curing are superior to that obtained by the closet prior art in low viscosity of the composition and improved mechanical properties of the cured article.

The undersigned declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

This 14th day of December, 2009

by Yoshikatsu Ichiryu

Yoshikatsu Ichiryu

We, the undersigned witnesses, hereby acknowledge that Yoshikatsu Ichiryu is personally known to us and did execute the foregoing Declaration in our presence on:

Date: December 14, 2009

Witness Nao Fujita

Date: December 14, 2009

Witness Yoshiyuki Kohno